Reply to Office action of 10/03/2005

IN THE UNITED STATES PATENT AND TRADEMARK OFFICE

Applicant:

Ji, J.

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Art Unit:

1723

Cust. No.:

02101 Examiner:

Fortuna, A.M.

Invention:

DEFECT FREE COMPOSITE MEMBRANES, METHOD FOR

PRODUCING SAID MEMBRANES AND USE OF THE SAME

#### Certificate of Mailing

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Barbara J. Carter, Ph.D

Mail Stop Amendment Commissioner for Patents PO Box 1450 Alexandria, VA 22313-1450

## DECLARATION OF JIANG JI, PH.D. IN SUPPORT OF APPLICANT'S RESPONSE [37 C.F.R. § 1.132]

Dear Sir:

In support of the accompanying response to the Office Action mailed October 03, 2005 in the above-reference matter, I hereby declare as follows:

1. My name is Jiang Ji, Ph.D. I am the inventor of the subject matter of the above patent application and am an inventor or co-inventor on a number of other patents and pending patent applications involving membrane technology, including U.S. Patent Nos. 6,596,167 and 6,890,435 and US Application Serial Nos. 10/604,664 and 10/857,531. I have a Ph.D. in Organic/Polymer Chemistry from McMaster University, Hamilton,

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Ontario, Canada and I am an internationally known scientist in the field of membrane science and technology and am an author or co-author of a number of publications involving membrane technology, including,

- (i) J. Ji, and M. Mehta, Mathematical Model for the Formation of Thin-Film Composite Hollow Fiber and Tubular Membranes by Interfacial Polymerization, J. Memb. Sci., 192, 41-54, (2001).
- (ii) J. Ji, J. M. Dickson, R. F. Childs, and B. E. McCarry, Mathematical Model for the Formation of Thin-Film Composite Membranes by Interfacial Polymerization: Porous and Dense Films, Macromolecules, 33, 624-633, (2000).
- (iii) J. Ji, B. J. Trunsinski, R. F. Childs, J. M. Dickson, and B. E. McCarry, Fabrication of Thin-Film Composite Membranes with Pendent, Photoreactive Diazoketone Functionality, Journal of Applied Polymer Science, 64, 2381-2398, (1997).
- (iv) J. Ji, R. F. Childs, M. Mehta, Mathematical Model for Encapsulation by Interfacial Polymerization, J. Memb. Sci., 192, 55-70, (2001).
- (v) J. Ji, M. Fei, G. Fan, J. Chen, G. Li, and W. Xiu, The Nucleation of Organic Additives in Membrane Casting Solution (I), The Theory and Direct Evidence, Desalination, 85, 297 -320, (1992).
- (vi) J. Ji, and J. Chen, Study on Interaction between PBIL Membranes and Organic Solutes, Desalination, 78, 389 396, (1990).
- (vii) J. Ji, M. Sun, M. Fei, and J. Chen, Study on the Interaction between Membranes and Organic Solutes by the HPLC Method, Desalination, 71, 107-126, (1989).

My further credentials are set forth in my Curriculum Vitae, which is attached as Exhibit A hereto.

2. I have read the action of October 3, 2005. This declaration is provided to distinguish the presently claimed membranes and methods of preparing and using such

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membranes from the cited art, and explain why the presently claimed invention is novel and non-obvious over the cited art.

- I have carefully reviewed your comments and the cited patents, and I realize that it is necessary to make some clarifications. I am ware of the prior arts you cited, they may appear to be related to my invention, but they do not provide teaching that will lead to my invention. In contrast, my invention has discovered novel membranes and novel methods for making said novel membranes to overcome the problems that the prior art was unable to solve or did not encounter at the time. My invention has significantly advanced the art of membrane fabrication processes and has produced novel membranes that show much more advanced properties than the prior art.
- The following is my detailed response to the Examiner's comments in the pending Office Action of October 3, 2005. First, I would like to explain the claimed invention from a chemical reaction perspective. Under the conditions utilized in the present invention, poly(vinyl butyral-co-vinyl-alcohol-vinyl acetate), a key ingredient of the membrane formulation in Examples 1 to 4 of the present invention, reacts with each other catalyzed by aluminum chloride, another key ingredient of the membrane formulation, to form cross-linked three dimensional network, which entangle with poly(vinylidene fluoride) (PVDF) network to strengthen the membrane in the present invention. In addition, aluminum chloride also reacts with poly(vinyl butyral-co-vinyl-alcohol-vinyl acetate) to form cross-linked poly(vinyl butyral-co-vinyl-alcohol-vinyl acetate) network having O-Al-O linkages. In this reaction, aluminum chloride plays a role of both reactant and catalyst. Poly(vinyl butyral-co-vinyl-alcohol-vinyl acetate) also reacts with polyester braid catalyzed by aluminum chloride to form ester linkage, acetal linkage and O-Al-O

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bridge to make the membrane permanently attach to polyester braid through covalent bonding.

- 5. The specially formulated adhesive used in Example 2 of the present invention forms a three-dimensional adhesive network, which further reacts with the braid and membrane to bond membrane and braid together through covalent bonding. This reaction is totally different from that disclosed in U.S. Patent 5,914,039, where both base and acid are needed in order to get the PVDF react. In the present invention, no acid is needed at all. Thus, the present invention is more advanced than the art disclosed in patent '039.
- 6. The adhesive is covalently bonded to the braid by ester and amide linkages; and the membrane is covalently bonded to the adhesive by carbon-carbon bond and ether linkage, i.e., the adhesive layer is sandwiched between the membrane and braid by covalent bonding as depicted in Fig. 2 of the present invention to form an adhesive reinforced composite membrane. This is clearly different from Fig. 3 of patent '039, where a single layer membrane made from the same polymer is inaccurately or incorrectly described as four layers, 35, 36,37 and 38. The further clarification is provided below regarding specific points raised by the Examiner in the Office Action.
- 7. A specially formulated adhesive used in Example 3 of the present invention completely penetrates through the braid support and partially penetrates into membrane layer from inside to covalently bond the braid and membrane together to form a defect free composite membrane. In other words, the specially formulated adhesive is covalently bonded with the braid by ester and amide linkages; and covalently bonded with the membrane by carbon-carbon bond and ether linkage. The chemical reaction of the adhesive used in Example 3 of the present invention is totally different from the

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membrane shown in Figs. 3-5 of patent '039, where there is no chemical binding at all between the membrane and braid.

- Fig. 3 of patent '039 shows a sketch of membrane cross section, it has multiple 9. regions with different porosity. Although the sketch in Fig. 3 of patent '039 looks somewhat similar to the sketch in Fig. 2 of the present invention, actually it is totally different after careful examination and comparison. In fact, in Fig. 3 of '039, elements 35, 36, 37, 38 are formed from the same polymer by coagulation, they are simply detailed porous structures formed from the same membrane casting solution as shown by a scanning electron photomicrograph in Fig. 4 of '039, the detail description is given in column 8, lines 21-39 of '039. Let me emphasize on this point by quoting column 8 lines 28-33 of '039: "... as evident in great detail in the photomicrograph FIG. 4. The skin is very thin, dense layer of polymer formed as dope contact the coagulant. By reason of manner in which the skin and each layer is formed from the same polymer, the layers have, in a radially inward direction from under the skin to the braided yarn 39 which defines the bore 32, progressively larger pores." This means that these layers, 35, 36, 37, 38 of '039 are formed from a single membrane casting solution in a single coagulation process at the same time, there is no difference in chemical composition between regions

.......

having different pore size and pore size distribution. The sketch shown in Fig. 3 of '039 is an incorrect representation of the true membrane structure shown in Figs. 4 and 5 of '039. Furthermore, '039 does not teach using covalent bonding between braid and coating layer to make a defect free membrane.

- 10. In contrast to the teaching of '039, the three layers shown in Fig. 2 of the present invention are made by coating two solutions in two sequential steps in a single spinneret shown in Fig. 1 of the present invention. The first coating 22 covers the rough surface of the braid 20 and provides sooth surface for second coating 21. On one hand, the first coating 22 provides covalent bonding with the braid 20 to anchor the coating 22 onto the braid 20 permanently; on the other hand, the first coating 22 also provides covalent bonding with the second coating 21 to form a composite membrane reinforced by covalent bonding within the adhesive layer; within the membrane layer; and between the braid and adhesive layer; as well as between the adhesive and membrane layers. Thus, the composite membranes of the present invention are not only structurally (physically) different, but also chemically different from those disclosed in patent '039.
- Patent '039 does not reveal membrane defect and adhesion problems between the braid and membrane nor the impact of braid physical structure on membrane performance, but US Pat. No. 6,354,444 (hereinafter patent '444), which is a continuation-in-part of patent '039, issued to the same group of inventors does indeed reveal the problems associated with membrane adhesion, braid physical structure, and pin-hole defects in columns 1 lines 25-67 and column 2 lines 1-36. A method to address these issues is disclosed by emphasis on using different physical structures of braids, Fig. 1 A-C, column 2 lines 49-67, column 3 lines 1-35 and claims 1-11 of '444.

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- 12. In the present invention, these critical problems alluded to in the previous paragraph, in particular, membrane defects, and insufficient membrane adhesion to the support, have been solved using chemical reaction approach and sequential multiple-layer coating in a single spinneret. The novel approach of the present invention has not been taught by the teaching of '039 nor '444, although a complex of PVDF with calcined α-alumina particles and polyvinyl alcohol are used to make braid support membranes. '039 and '444 do not teach using covalent bonding and multiple coatings to strengthen adhesion and to eliminate membrane defects. Instead, '444 chooses the physical approach using different braid structure to improve membrane adhesion, this physical approach of '444 is far less effective than the chemical approach of the present invention. Thus, the present invention provides more advanced membranes than the prior art in terms of membrane integrity, selectivity and flux.
- 13. The Examiner asserts that "[c]laim 2 is also disclosed (column 8, lines 23-34)." See Office Action, p. 2. As discussed above, the second half of claim 2 of the present invention, i.e. "The membrane of claim 1, wherein said middle layer and said outside barrier layer are formed from.........different coating solutions" is clearly different from those described in column 8, line 23-34 of '039, where the so-called "layers" are actually different porous regions of the same single layer membrane formed from the same polymer. (column 8, lines 31-32). In other word, the '039 authors incorrectly or inaccurately described the different porous regions of a materially same single layer membrane, which real structure is shown in Figs. 4 and 5 of '039, as a multiple-layer membrane in Fig. 3 of patent '039.

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- In contrast to '039, the membrane claimed in the second half of claim 2 of the present invention is a true materially different multiple-layer membrane, each layer has distinct chemical composition as described in Example 2 of the present invention, where the first layer is specially formulated adhesive, the second membrane layer is made of dope I consisting of poly(vinyl butyral-co-vinyl-alcohol-vinyl acetate), aluminum chloride, polyvinylpyrrolidone, and poly(vinylidene fluoride), which form three dimensional network to strengthen the membrane in the present invention.
- 15. In addition, aluminum chloride also reacts with poly(vinyl butyral-co-vinyl-alcohol-vinyl acetate) to form crosslinked poly(vinyl butyral-co-vinyl-alcohol-vinyl acetate) network having O-Al-O linkages. The membrane obtained from Example 2 of the present invention is schematically illustrated in Fig. 2, which is a true multiple-layer membrane. The polyester braid provides mechanical support; PVDF entangled with three dimensional network of crosslinked poly(vinyl butyral-co-vinyl-alcohol-vinyl acetate) forms a barrier layer to provide selective separation; and middle adhesive layer formed from specially formulated adhesive, bonds the outer barrier layer and inner braid support together to form an reinforced composite membrane. The middle adhesive layer of the present invention is different from the conventional adhesive in that the middle adhesive layer of the present invention is permeable to fluid; while the conventional adhesive is not permeable to fluid, it is normally used to seal membrane when making a membrane cartridge.
- 16. The first half of claim 2 of the present invention, i.e. "The membrane of claim 1, wherein said middle layer and said outside barrier layer are formed from the same coating solution" is also different from the those described in column 8, line 3-34 of patent '039

in that the chemical composition of the present invention is different from that of '039. In the present invention, poly(vinyl butyral-co-vinyl-alcohol-vinyl acetate) is used as a key ingredient of the membrane, which reacts with each other catalyzed by aluminum chloride to form cross-linked three dimensional network through covalent bonding of acetyl bond linkage and O-Al-O linkages. Cross-linked three dimensional network of poly(vinyl butyral-co-vinyl-alcohol-vinyl acetate) entangles with PVDF network to strengthen the membrane in the present invention.

- 17. In addition, aluminum chloride also reacts with poly(vinyl butyral-co-vinyl-alcohol-vinyl acetate) to form O-Al-O bridge to crosslink multiple poly(vinyl butyral-co-vinyl-alcohol-vinyl acetate) macromolecules together. In this reaction, aluminum chloride plays a role of both reactant and catalyst. More importantly, poly(vinyl butyral-co-vinyl-alcohol-vinyl acetate) reacts with polyester braid catalyzed by aluminum chloride to form ester linkage, acetal linkage and oxygen-aluminum-oxygen bridge to make the membrane permanently attach to polyester braid. In this reaction, aluminum chloride again plays a role of both reactant and catalyst.
- 18. Therefore, the defect free composite membranes claimed in claims 1 and 2 are totally different from those disclosed in patent '039 in terms of both chemical composition and physical structure, thus giving much higher flux than those disclosed in the '039 patent which have similar retention characteristics. Thus, the membranes of the present invention have much more advanced features than the prior art.
- 19. With respect to the Examiner's assertion that "RE: claim 3, the middle layer is formed from organic polymer, e.g. hydrophilic PVDF (column 7, second paragraph)"

(see Office Action, p. 3), there are at least two major differences between the claim 3 of the present invention and what described in column 7, second paragraph of patent '039.

- (1) Claim 3 of the present invention emphasizes that the middle layer, "has excellent compatibility between the support and the barrier layers to bond them together." The prior art described in column 7, second paragraph does not mention to bond the membrane with the braid support at all. In other word, patent '039 does not teach bond the membrane with the support. As pointed out above, patent '039 does not reveal membrane defect and adhesion problems between the braid and membrane nor the impact of braid physical structure on membrane performance, but US Pat. No. 6,354,444 (hereinafter patent '444) issued to the same group of inventors does indeed reveal the problems associated with membrane adhesion, braid physical structure, and pin-hole defects in columns 1 lines 25-67 and column 2 lines 1-36.
- 20. A method to address these issues is disclosed by emphasis on using different physical structures of braids, Fig. 1 A-C, column 2 lines 49-67, column 3 lines 1- 35 and claims 1-11 of '444. In contrast to the teaching of '039, claim 3 of the present invention describe the bonding membrane with the support by adhesives and other organic and inorganic materials which have bonding capacity similar to expoxy, polyurethane, silicone and other adhesives.
- 21. (2) Claim 3 of the present invention describes the middle layer of the membrane is selected from the group of consisting of expoxy, polyurethane, silicone and other adhesives, including other organic and inorganic materials which have bonding capacity similar to adhesives. The prior art described in column 7, second paragraph does not teach using adhesive at all, instead describes PVDF/α-Aluminum complex, which does

not provide bonding between the membrane and support as revealed by the same group of inventor in a subsequent US Patent No. 6,354,444. Therefore, the prior art described in column 7, second paragraph of patent '039 does not teach claim 3 of the present invention. In contrast, claim 3 of the present invention claims much more effective composition of matters than the prior art of '039 to bond chemically and physically membrane and support together.

22. With respect to the Examiner's comment that: "Re: claims 4-6, and 8, the membrane in tubular and or hollow fiber shape is disclosed (column 11, lines 39-54, claim 1)," (see Office Action, p. 3), I am aware that the hollow fiber and tubular membranes existed even before patent '039. As described above, the composite membranes described in Examples 1-4, in particular, Examples 2 and 3 of the present invention have covalent bonding between the membrane and braid support, and between the adhesive layer and membrane. These covalent bonding within the layer and between the layers strengthen the membrane and significantly improve overall membrane performance in terms of integrity, flux and separation. The present invention has eliminated the membrane defects and delamination problems that patent '039 fails to solve. As you can see from the above clarification, patent '039 does not teach what I invented in the present invention, even for people who are skilled at art can not get a clue from patent '039, which is evident in patent '444, a physical approach using different braiding pattern was chosen, not a chemical approach as described in the present invention. The novelty and inventiveness of the present invention to eliminate membrane defect and to overcome membrane delamination problem by double layers coated one on top of another in sequence in a single spinneret; and by covalent bonding between the

support and coating layers is acknowledged by the Written Opinion of the International Searching Authority for the related PCT application (PCT/US04/25374), a copy of which is attached herewith as Exhibit B.

- 23. With respect to the Examiner's comment that: "Re: claim 9, the claimed membrane properties are disclosed in column 10, line 45-56)" (see id), I am aware that the '039 membrane is used for water filtration, but its highest specific flux is only 36.6 gfd/psi. In contrast, the membrane of the present invention has the specific water flux as high as 141 gfd/psi, which is 300% higher that of '039. Thus, the membranes of the present invention have much more advanced properties than those disclosed in the prior art of patent '039.
- 24. Claim Rejections-35 USC§ 103(a)

Claim 7 is rejected as being unpatentable over the '039 patent in further view of Strobel et al (5,766,473 – hereinafter the '473 patent). Coating spheres is not new. What is new in the present invention is coating spheres with multiple reactive coating layers to give a unique spherical defect free membrane, which spheres have chemical composition and separation properties similar to its hollow fiber and flat sheet counterparts, but different from those disclosed in the prior art.

25. In patent '039, the calcined alumina particle size (<5 micrometer) is much smaller than the thickness of film. The particles are used as film fillers, and are suspended in the casting solution, this is different from coating a sphere, in this case, the thickness of coating layer is normally much smaller than the diameter of sphere. The volume ratio of coating layer to coated particle is extremely small in the present invention, thus the properties associated with this ratio is quite different from that of patent '039, where the

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volume ratio of coating layer to coated particles is huge, the thickness of the coating is about 20 times larger than the size of calcined alumina particle.

- 26. US Pat. No. 5,766,473 discloses the coating of thermally formed polyethylene and polypropylene microfiltration membranes with tactic hydrophilic poly(vinyl alcohol). On micro scale, polyethylene and polypropylene membrane have microporous structure, the fibrous polyethylene and polypropylene are connected by randomly shaped knots to form three dimensional network. Tactic poly(vinyl alcohol) covers these fibrous knots to form a shell around the outer surface. Enzymes as oxygen scavenger are loaded onto the poly(vinyl alcohol) shell. On macro scale, the membranes of patent '473 have flat sheet geometry.
- 27. The major differences between the present invention and patents '473 and '039 are as follows: In the present invention, there are three different scenarios:
  - (1) for a spherical support which has reactive groups, such as esters, the support is first coated by a reactive casting solution, which reacts with the support to form covalent bonding to permanently bond the coating layer to the support; such covalently bonded first coating layer is coated with a second reactive layer that reacts with the first coating layer to form a defect free composite membrane with covalent bonding and cross-linking to strengthen the defect free composite membrane. As described above, in the present invention, poly(vinyl butyral-covinyl-alcohol-vinyl acetate) is used as a key ingredient of the membrane, which reacts with each other catalyzed by aluminum chloride to form cross-linked three dimensional network through covalent bonding of acetyl bond linkage and O-Al-O linkages. Cross-linked three dimensional network of poly(vinyl butyral-co-

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vinyl-alcohol-vinyl acetate) entangles with PVDF network to strengthen the membrane in the present invention. In addition, aluminum chloride also reacts with poly(vinyl butyral-co-vinyl-alcohol-vinyl acetate) to form O-Al-O bridge to crosslink multiple poly(vinyl butyral-co-vinyl-alcohol-vinyl acetate) macromolecules together. In this reaction, aluminum chloride plays a role of both reactant and catalyst. More importantly, poly(vinyl butyral-co-vinyl-alcohol-vinyl acetate) reacts with polyester braid catalyzed by aluminum chloride to form ester linkage, acetal linkage and oxygen-aluminum-oxygen bridge to make the membrane permanently attach to polyester braid. In this reaction, aluminum chloride again plays a role of both reactant and catalyst.

- (2) For a spherical support, which does not have obvious reactive groups, the support is first coated by a reactive adhesive layer which is not cured or only partially cured during coating, and the adhesive penetrates into the support. This reactive first coating layer is coated again with a second reactive layer, which reacts with the first coating layer to form a defect free composite membrane strengthened by covalent bonding and cross-linking within and between the layers.
- (3) For a support which has reactive groups, such as esters, the support is first coated by a reactive adhesive layer which is not cured or partially cured during coating, the reactive adhesive reacts with ester group to form covalent, the adhesive also penetrates into the support and reacts with each other to form cross-linked first coating layer which is covalently anchored onto the support, which is coated again with a second reactive layer, which reacts with the first coating layer

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to form a defect free composite membrane strengthened by covalent bonding and cross-linking.

- 28. Thus, the spherical membranes of the present invention are structurally (physically) and chemically different from those of the prior art. The present invention emphasizes on using reactive multiple coating layers, one layer on top of another, covalent bonding and cross-linking to make defect free composite membranes which have much higher flux, integrity and selectivity than those membranes disclosed in patent '193. The membranes disclosed in patent '473 are for oxygen removal. This is totally different from the present invention. Thus, the combination of patent '039 with patent '473 and patent '193 does not provide teaching for the present invention.
- 29. Regarding the Examiner's assertion in point 5 that "[c]laims 2 and 4 are rejected under 35 U.S.C. 103 (a)..." (see Office Action, p. 4), patent '039 fails to disclose the coating solutions of different polymers, as claimed in a second embodiment of claim 2. Careful examination of patent '039 indicates that patent '039 actually discloses coating solution of different polymers. In example 1, column 11, line 1 to 36 of patent '039, the detail formulation of coating solution of different polymers and dope preparation procedure are described. In example 2, column 11 line 42 it states that "the dope formed in Example 1 is fed to a nozzle ...". US Patent No. 6,024,872, which is a division of patent '039, also discloses the detailed formulation of coating solutions of different polymers and dope preparation procedures.
- 30. The Examiner's asserts that "[p]atent '193 teaches a braided support impregnated and coated with a first polymer, coating the support to form a layers on the braided polymeric material, and further using the coated support to canst a film on a additional

polymeric membrane, to increase the thickness of the membrane is disclosed (see entire disclosure, in particular claims 1, 8-11 and 21)." See Office Action, pp. 4-5. After having carefully examined patent '193 and entire disclosure, in particular claims 1, 8-11 and 21, I found that it is necessary to clarify the difference between the present invention and patent '193.

- 31. The major differences between the present invention and patent '193 are as follows. In the present invention, there are three different scenarios:
  - (1) for a support, which has reactive groups, such as esters, the support is first coated by a reactive casting solution, which reacts with the support to form covalent bonding to permanently bond the coating layer to the support; such covalently bonded first coating layer is coated with a second reactive layer that reacts with the first coating layer to form a defect free composite membrane with covalent bonding and cross-linking to strengthen the defect free composite membrane.
  - (2) for a support, which do not have obvious reactive groups, the support is first coated by a reactive adhesive layer which is not cured or partially cured during coating, the adhesive penetrates into the support. The first reactive coating layer is coated again with a second reactive layer, which reacts with the first coating layer to form a defect free composite membrane strengthened by covalent bonding, cross-linking and covalently anchoring onto the support.
  - (3) for a support, which has reactive groups, such as esters, the support is first coated by a reactive adhesive layer. Before the first reactive coating layer is cured, it is coated again with a second reactive layer, which reacts with the first

coating layer to form a defect free composite membrane firmly anchored onto the support by covalent bonding and cross-linking.

- 32. Thus, the present invention emphasizes on using reactive multiple coating layers, one layer on top of another, covalent bonding and cross-linking to make defect free composite membranes which have much higher flux, integrity and selectivity than those membranes disclosed in patent '193.
- 33. In patent '193, column 8 lines 24-31 it states: "Any type of resinous material for the supporting tube either alone or in combination with the other fibrous support may be used. Some suitable resins which may be used include phenolics, epoxies, polyurethanes, polyesters, acrylics, etc., or additionally thermoplastic resins, such as olefinic resin such as polyethylene, polypropylene, polyvinyl chloride, polystyrene etc., may also be used." These resins including epoxies and polyurethanes are fully cured resins before coating the membrane, thus do not undergo chemical reactions under the conditions used in patent '193. Therefore, there is no chemical bonding between the membrane and support.

  Instead, the membrane is physically attached to the support as described in patent '193, column 3 lines 67-71 where it states: "In preferred embodiment sufficient casting solution to completely penetrate the fabric. Complete penetration of the supporting material provides the greatest degree of support and adhesion."
- 34. Although patent '193 describes the use of epoxies and polyurethanes as support materials, it does not teach strengthening the adhesion between membrane and support by covalent bonding and cross-linking. In patent '193, the pin-hole problem caused by the short fiber ends projecting from the braid surface remain. On the other hand, this problem has been successfully resolved in the present invention by coating the support with two

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consecutive coatings in a single spinneret, covalent bonding and cross-linking further strengthen the defect free composite membrane.

- 35. In patent '193 claim 1, it states "(b) casting on to one surface of braided material a liquid film-forming polymeric composition ....; (c) impregnating the braded material with said polymeric composition to form a smooth thin-film layer on the cast surface of the material;" That means that there is only one coating layer, which is first cast onto the surface of the braid material; followed by impregnating the braid with same said polymeric composition. There is no second coating layer, the cast polymeric composition simply impregnates the braid after casting by penetrating into the braided material as described in Claim 4.
- 36. Claims 8-11 and 21 in '193 describe the same process as described above, i.e. "casting and impregnating the braided material with said polymeric composition....." as stated in claim 9. Claim 8 describes the detail process of casting and impregnating the inner surface of the braided material with said polymeric composition, while claim 9 describes "casting and impregnating the braided material with said polymeric composition on the outer surface of tubular braided material." Claims 8 and 9 describe casting polymeric composition onto inner and outer surface of the tubular braided material in two separated cases as shown in Fig. 4 and 5 of patent '193, respectively. This is two separate processes these casting processes are not in sequence, i.e., not one after another and not reactive during and after coating under the conditions disclosed and there is only one coating layer in each case.
- 37. Claim 10 describes a fabric material, which has double-layered braid strand, to provide a smooth surface on both side of strands, there is no coating at all. Claim 11

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describe the monofilament that is used to make braided material, again there is no coating at all. Claim 21 describes "...casting a fluted thin layer of the liquid film-forming polymeric composition onto the wall of the supported materials; and gelling the cast layer to form an integrally supported membrane..." There is only one coating here, there is no second coating.

- 38. However, claim 7 in patent '193 describes "the method of claim 1, which includes, prior to casting and impregnating steps, the wetting of surface of tubular braided material with a liquid." The reason for wetting the braid with liquid, (column 4 lines 18-51), is to form "a tubular configuration, for example, around a glass tube" for making a tubular membrane. Patent '193 does not mention improving adhesion between the membrane and support by covalent bonding at all, although "the monofilaments are resin-impregnated glass fibers" in claims 12 and 20 and "resin-impregnated nylon fiber" in claim 13. The reason to use resin-impregnated glass fibers and resin-impregnated nylon fibers is that these resin-impregnated fibers are "essentially free of short fiber ends which project from the surface of the strands." The "essentially free" does not means completely free, indeed, it is occasionally found short fiber ends which project from the surface of the strands to cause pin-hole defects of membrane.
- 39. In present invention, such short fiber ends, which project from the surface, are coated with a first coating solution to force the projected fiber ends attach to the braid surface, which is covered again by a second coating solution to give a defect free composite membrane. This is one of the major differences between the current invention and prior art. More importantly, patent '193 does not teach providing adhesion between membrane and support by using adhesive and covalent bonding at all. Instead, patent

'193 teaches physically impregnating braid with a membrane casting solution as described in column 3 lines 67-71: "In preferred embodiment sufficient casting solution to completely penetrate the fabric. Complete penetration of the supporting material provide the greatest degree of support and adhesion." The impregnating in patent '193 simply means that the casting solution physically penetrating braid, no chemical bonding is mentioned at all. The statement in column 3 lines 62-64: "The level of impregnation of the porous support .................. is not critical.", is not correct, it also contradicts to the statement in column 3 lines 67-71 of patent '193.

- 40. More importantly, the tubular membranes claimed in patent '193 are prepared by a non-continuous batch mode at very slow coating speed, the longest tube disclosed in '193 is 44 ft long. In contrast, the membrane of the present invention is made in a continuous process at a much higher speed, the membrane can be made from a few inch long to many miles long, i.e. as long as needed, because the process can be operated, 24 hours a day, 7 days a week, around the clock.
- 41. It should be pointed out that the membranes of the present invention not only have different chemical compositions from those of the '193, but also have much better performance than those of '193. For example, the membranes disclosed in patent '193 have extremely low flux, such as 20 gfd at 10 psi transmembrane pressure (column 5, line 2-26) which is 70 times lower than the membrane flux of the present invention, which is as high as 141 gfd/psi.
- 42. Therefore, after the teaching of patent '193 (even if combined with patent '039 and patent '473), even the people who are skilled in the art cannot obtain the membranes of the present invention. The membranes of the present invention show much more

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advanced properties than those of the prior art in terms of flux, selectivity and integrity.

Furthermore, the membrane fabrication processes of the present invention are much more productive than those disclosed in the prior art in terms of coating speed and mass transfer efficiency, because ultrasonic waves are utilized to promote phase inversion and solvent removal from the membrane.

- different from those disclosed in the patents cited by the examiner. In the present invention, the adhesives including epoxy, polyurethane, silicones react with both membrane and support to covalently bond the membrane and support together to form a support reinforced membranes, which are much stronger, much more permeable, and much more selective than those disclosed in prior art. More importantly, the prior art does not provide teaching that will lead to the present invention.
- 44. The coating processes of the present invention for making defect free composite membranes are much more and advanced than those disclosed in prior art, in particular, patents '039 and 193. The coating processes of patent '193 are manual processes. The tubular membranes claimed in patent '193 are prepared by a non-continuous batch mode at very slow coating speed, the longest tube disclosed in '193 is 44 ft long. The coating process of patent '039 are described in column 3 lines 51-64, and in column 11 lines 40-60. A tubular braid is coated with a layer of dope containing calcined α-alumina particles, polyvinyl alcohol and PVDF at a speed of 40 ft/min, coagulated in water to form a braid support membrane. No ultrasonic wave stirring is used to facilitate phase inversion.
- 45. In contrast, the membrane of the present invention is prepared by coating a braid with two reactive layers simultaneously in a single spinneret, one layer on top of the

Appl. No. 10/620,715

Amdt. Dated 12/23/05

Reply to Office action of 10/03/2005

other, at a coating speed as high as 100 ft/min or higher. The ultrasonic wave generators are used to provide ultrasonic agitation in coagulation bath to facilitate mass transfer between cast membrane and coagulation liquid to speed up phase inversion from liquid to solid of the cast membrane. Thus, the membranes are coated at much higher speed in the present invention than prior art. The membranes in the present invention are produced in a continuous mode. The membranes are collected by two interchangeable membrane take-up wheels, when one is full, switching to another, the process can be operated around the clock, 24 hours a day, 7 days a week. The coating processes of the present invention are much more efficient and much more productive those disclosed in the prior art.

- 46. The above discussion clearly shows that the novel membranes and novel processes for making said novel membranes of the present invention are much more advanced than those disclosed in prior art. The prior art does not provide teaching that will lead to the present invention. Thus, even the people who are skilled at art cannot develop of the same membranes and processes of the present invention by following the teaching of the prior art.
- 47. I hereby declare that all statements made herein are of my own knowledge and that all statements made on information and belief are true; and further that these statements are being made with the knowledge that willful false statements and the like so made are punishable by fine or imprisonment or both under Section 1001 of Title 18 of the United States Code and that such willful false statements may jeopardize the validity of the application or any patent issued thereon.

22

Appl. No. 10/620,715 Amdt. Dated 12/23/05 Reply to Office action of 10/03/2005

Respectfully yours,

Jiang Ji

Tel: (603) 560-2147 Fax: (603) 458-1483

Email: jijiang@comcast.net

4 Hawk Drive Salem, NH 03079

Dated: December 23, 2005



#### Exhibit A

## Jiang Ji, Ph.D.

Member of American Chemical Society Member of North America Membrane Society

**EXPERIENCE** 

June 2004 - Present

**Staff Scientist** 

Amersham Biosciences, GE Healthcare

General Electric Company Westborough, MA 01581

Research and development of cutting edge technology and products for purification of cervical cancer vaccine

Sept. 1998 - May, 2004

**Senior Polymer Scientist** 

Koch Membrane Systems Inc., Koch Industries, Inc.

Wilmington, MA, USA

Research and development of reverse osmosis (RO), nanofiltration (NF), ultrafiltration (UF), and microfiltration (MF) membranes having different geometries, ranging from hollow fiber, tubular to flat sheet membranes for water purification, wine filtration, juice filtration, and other applications in biotech, pharmaceutical, food and chemical industries

Sept. 1995 - Sept. 1998

Senior Research Chemist

Zenon Environmental Inc., ON, CANADA

Research and development of braid reinforced hollow fiber membranes and tubular membranes for drinking water purification, wastewater treatment and other applications in auto, food, biotech, pharmaceutical and chemical industries.

March - Aug. 1989

Visiting Research Scientist University of Oxford, UK

Study of natural phospholipid vesicles/transition metal catalysts

Sept. 1988 - Feb. 1989

Visiting Research Scientist University of Ottawa, CANADA

Study on reverse osmosis membranes and nanofiltration membranes

June 1986 - Aug. 1988

**Research Scientist** 

**Dalian Institute of Chemical Physics** 

**Chinese Academy of Sciences** 

Fundamental research on membrane formation, membrane transport mechanism and interactions

between membrane and separated solutes

July 1985 - May 1986

Lecturer

**Dalian University of Technology** 

Teaching physical chemistry and chemical engineering

**EDUCATION** 

Sept. 1989 - Aug. 1995

Ph.D. Organic/Polymer Chemistry

**Department of Chemistry** 

McMaster University, Hamilton, ON, CANADA

Ph.D. Dissertation:

Fabrication and Photochemical Surface Modification of Photoreactive Thin-Film Composite Membranes and Model Development for Thin Film Formation by

**Interfacial Polymerization** 

Sept. 1982 - July 1985

M.Eng.

Department of Chemical Engineering

**Dalian University of Technology** 

M.Eng. Thesis:

Catalytic Characteristics of the Keggin Type of

Heteropolyacids

March 1978 - Jan. 1982

B.Eng.

**Department of Chemical Engineering** 

**Dalian University of Technology** 

**B.Eng. Thesis:** 

Synthesis of poly(ethylene-co-butadiene) using Ziegler-

Natta catalyst and rare earth metal catalysts

Exhibit B

## PATENT COOPERATION TREATY

Profit the INTERNATIONAL SEARCHING AUTHORITI	W <sub>D</sub>			
To: JIANG Л	PCT  NOTIFICATION OF TRANSMITTAL OF THE INTERNATIONAL SEARCH REPORT AND			
4 HAWK DRIVE SALEM, NH 03079	NOTIFICATION OF TRANSMITTAL OF			
SALEW, NR 03079	THE INTERNATIONAL SEARCH REPORT AND			
	THE WRITTEN OPINION OF THE INTERNATIONAL SEARCHING AUTHORITY, OR THE DECLARATION			
·	(PCT Rule 44.1)			
	Date of mailing (day/month/year)			
Applicant's or agent's file reference 000037663	FOR FURTHER ACTION See paragraphs I and 4 below			
International application No. PCT/US04/22502	International filing date (day/month/year) 14 July 2004 (14.07.2004)			
Applicant JI, JIANG	·			
1. The applicant is hereby notified that the internation search	d the written opinion of the International Searching Authority			
have been established and are transmitte				
Filing of amendments and star of u '9: The applicant is entitled, if he es la	antional ap tion (see Rule 46):			
When? The time limit !	ths from late of transmittal of the intentional			
search report.	-tamba			
Where? Directly to the 1211 Geneva 26	olombet.			
For more detailed instruc	Facility (1)			
2. The applicant is hereby notifi  Article 17(2)(a) to that effect and the written opinion of the	tablished that the declaration to the International Searching Additional Searching in the International Searching Additional Searching Additional Search Sea			
<b>}</b>	tional fee(s) under Rule 40.2, the applicant is notified that:			
the protest together with the decision thereon has be request to forward the texts of both the protest and t	en transmitted to the International Bureau together with the applicant's			
	plicant will be notified as soon as a decision is made.			
4. Reminders				
Bureau. If the applicant wishes to avoid or postpone publication, claim, must reach the International Bureau as provided in Rules	te, the international application will be published by the International a notice of withdrawal of the international application, or of the priority 90bis.1 and 90bis.3, respectively, before the completion of the technical			
preparations for international publication.	the written opinion of the International Searching Authority to the			
International Bureau. The International Bureau will send a cor	by of such comments to all designated Offices unless an international.  These comments would also be made available to the public but not			
before the expiration of 30 months from the priority date.				
Within 19 months from the priority date, but only in respect of some designated Offices, a demand for international preliminary examination must be filed if the applicant wishes to postpone the entry into the national phase until 30 months from the priority date (in some Offices even later), otherwise, the applicant must, within 20 months from the priority date, perform the prescribed acts for entry				
into the national phase before those designated Offices.  In respect of other designated Offices, the time limit of 30 months (or later) will apply even if no demand is filed within 19 months.				
	applicable time limits, Office by Office, see the PCT Applicant's Guide,			
Volume II, National Chapters and the WIPO Internet site.				
Name and mailing address of the ISA/ US	Authorized officer			
Mail Stop PCT, Attn: ISA/US Commissioner for Patents	Ana M. Fortuna			
P.O. Box 1450 Alexandria, Virginia 22313-1450	Telephone No. (571) 272-1700			
Facsimile No. (703) 305-3230				
Form PCT/ISA/220 (January 2004)				

## PATENT COOPERATION TREATY

## From the INTERNATIONAL SEARCHING AUTHORITY

To: JIANG JI	PCT				
4 HAWK DRIVE SALEM, NH 03079	NOTIFICATION OF TRANSMITTAL OF THE INTERNATIONAL SEARCH REPORT AND THE WRITTEN OPINION OF THE INTERNATIONAL SEARCHING AUTHORITY, OR THE DECLARATION				
	(PCT Rule 44.1)				
	Date of mailing (day/month/year) 24 OCT 2018				
Applicant's or agent's file reference 000037663	FOR FURTHER ACTION See paragraphs 1 and 4 below				
International application No. PCT/US04/22502	International filing date (day/month/year) 14 July 2004 (14.07.2004)				
Applicant JL JIANG					
The applicant is hereby notified that the international sear have been established and are transmitted herewith.	rch report and the written opinion of the International Searching Authority				
Filing of amendments and statement under Article 19: The applicant is entitled, if he so wishes, to amend the cla	: nims of the international application (see Rule 46):				
When? The time limit for filing such amendments is search report.	s normally two months from the date of transmittal of the international				
Where? Directly to the International Bureau of WIP( 1211 Geneva 20, Switzerland, Facsimile No	Where? Directly to the International Bureau of WIPO, 34 chemin des Colombettes 1211 Geneva 20, Switzerland, Facsimile No.: (41-22) 338.82.70.				
For more detailed instructions, see the notes on the	. 1				
2. The applicant is hereby notified that no international seal Article 17(2)(a) to that effect and the written opinion of t	rch report will be established and that the declaration under the International Searching Authority are transmitted herewith.				
	ditional fee(s) under Rule 40.2, the applicant is notified that:				
the protest together with the decision thereon has b	een transmitted to the International Bureau together with the applicant's the decision thereon to the designated Offices.				
no decision has been made yet on the protest; the a	pplicant will be notified as soon as a decision is made.				
4. Reminders  Shortly after the expiration of 18 months from the priority date, the international application will be published by the International Bureau. If the applicant wishes to avoid or postpone publication, a notice of withdrawal of the international application, or of the priority claim, must reach the International Bureau as provided in Rules 90bis.1 and 90bis.3, respectively, before the completion of the technical preparations for international publication.					
The applicant may submit comments on an informal basis on the written opinion of the International Searching Authority to the International Bureau. The International Bureau will send a copy of such comments to all designated Offices unless an international preliminary examination report has been or is to be established. These comments would also be made available to the public but not before the expiration of 30 months from the priority date.					
Within 19 months from the priority date, but only in respect of some designated Offices, a demand for international preliminary examination must be filed if the applicant wishes to postpone the entry into the national phase until 30 months from the priority date (in some Offices even later); otherwise, the applicant must, within 20 months from the priority date, perform the prescribed acts for entry into the national phase before those designated Offices.					
In respect of other designated Offices, the time limit of 30 months (or later) will apply even if no demand is filed within 19 months.					
See the Annex to Form PCT/IB/301 and, for details about the applicable time limits, Office by Office, see the PCT Applicant's Guide, Volume II, National Chapters and the WIPO Internet site.					
Name and mailing address of the ISA/ US	Authorized officer				
Mail Stop PCT, Attn: ISA/US Commissioner for Patents	Ana M. Fortuna				
P.O. Box 1450 Alexandria, Virginia 22313-1450 Facsimile No. (703) 305-3230	Telephone No. (571) 272-1700				

Form PCT/ISA/220 (January 2004)

(See notes on accompanying sheet)

## PATENT COOPERATION TREATY

## **PCT**

## INTERNATIONAL SEARCH REPORT

(PCT Article 18 and Rules 43 and 44)

Applicant's or agent's file reference 000037663	FOR FURTHER see Form PCT/ISA/220 ACTION as well as, where applicable, item 5 below.				
International application No. International filing date (day/month/year) (Earliest) Priority Date (		(Earliest) Priority Date (day/month/year) 16 July 2003 (16.07.2003)			
PCT/US04/22502 14 July 2004 (14.07.2004) 16 July 2003 (16.07.2003)					
Applicant Л. ЛАNG					
This international search report has been prepared by this International Searching Authority and is transmitted to the applicant according to Article 18. A copy is being transmitted to the International Bureau.					
This international search report consists of a total of sheets.  It is also accompanied by a copy of each prior art document cited in this report.					
	d by a copy of cach prior art document over				
<ol> <li>Basis of the Report         <ul> <li>With regard to the language, the</li> </ul> </li> </ol>	international search was carried out on the ba	sis of:			
	application in the language in which it was fil-				
a translation of the international application into, which is the language of a translation furnished for the purposes of international search (Rules 12.3(a) and 23.1(b))					
	ide and/or amino acid sequence disclosed in				
	d unsearchable (See Box No. II)				
3. Unity of invention is lacki					
4. With regard to the title,					
the text is approved as sub-	mitted by the applicant.				
the text has been established by this Authority to read as follows:					
		+			
·					
5. With regard to the abstract,		·			
the text is approved as sub					
the text has been establish may, within one month fro	ed, according to Rule 38.2(b), by this Authorit m the date of mailing of this international sear	y as it appears in Box No. IV. The applicant ch report, submit comments to this Authority.			
6. With regard to the drawings,					
. —	e published with the abstract is Figure No. 2				
as suggested by the		A . 6			
· —	s Authority, because the applicant failed to sug	The state of the s			
	s Authority, because this figure better character	rizes the invention.			
b none of the figures is to be published with the abstract.					

Form PCT/ISA/210 (first sheet) (April 2005)

## INTERNATIONAL SEARCH REPORT

International application No.

PCT/US04/22502

Box No. II Observations where certain claims were found unsearchable (Continuation of item 2 of first sheet)			
This international search report has not been established in respect of certain claims under Article 17(2)(a) for the following reasons:			
Claims Nos.: because they relate to subject matter not required to be searched by this Authority, namely.			
Claims Nos.:  because they relate to parts of the international application that do not comply with the prescribed requirements to such an extent that no meaningful international search can be carried out, specifically:			
3. Claims Nos.: because they are dependent claims and are not drafted in accordance with the second and third sentences of Rule 6.4(a).			
Box No. III Observations where unity of invention is lacking (Continuation of item 3 of first sheet)			
This International Searching Authority found multiple inventions in this international application, as follows:			
See USPTO299 (ATTACHED).			
<ol> <li>As all required additional search fees were timely paid by the applicant, this international search report covers all searchable claims.</li> <li>As all searchable claims could be searched without effort justifying additional fees, this Authority did not invite payment of any additional fees.</li> <li>As only some of the required additional search fees were timely paid by the applicant, this international search report covers only those claims for which fees were paid, specifically claims Nos.:</li> </ol>			
4. No required additional search fees were timely paid by the applicant. Consequently, this international search report is restricted to the invention first mentioned in the claims; it is covered by claims Nos.: 1-9  Remark on Protest  The additional search fees were accompanied by the applicant's protest and, where applicable, the payment of a protest fee.  The additional search fees were accompanied by the applicant's protest but the applicable protest fee was not paid within the time limit specified in the invitation.  No protest accompanied the payment of additional search fees.			

#### INTERNATIONAL SEARCH REPORT

International application No.

PCT/US04/22502

Box IV TEXT OF THE ABSTRACT (Continuation of Item 5 of the first sheet)

The technical features mentioned in the abstract do not include a reference sign between parentheses (PCT Rule 8.1(d)).

#### **NEW ABSTRACT**

A defect free semipermeable membrane ((Fig. 2)) having excellent integrity and high water permeability is provided. Said composite membrane comprises an inside support layer (20) to provide sufficient mechanical strength, and outside barrier layer (21) to provide selective separation and a middle layer (22) to provide both chemical and physical binding between the support and the barrier layers. Three different methods for making said defect free are disclosed. These methods have been successfully utilized to produce high quality coatings and defect free composite membranes, which are independent of chemical composition and physical structure of said support. In the present invention, the ultrasonic sonification is utilized to enhance mass transfer and to speed up the phase inversion process of a membrane casting solution, and to produce a composite membrane at a speed higher than those disclosed in the prior art. Said defect free composite membrane have broad applications, ranging from filtration of fruit juice, wine, and milk to purification of drinking water, and municipal and industrial water.

Form PCT/ISA/210 (continuation of first sheet(3)) (April 2005)

## INTERNATIONAL SEARCH REPORT

International application No.

PCT/US04/22502

A. CLASSIFICATION OF SUBJECT MATTER  IPC(7): BO1D 61/00, 63/00  US CL: 210/490, 500.27, 500.35, 500.41, 500.36, 500.42; 264/41, 216; 428/310.5; 96/4, 10, 11; 95/45  According to International Patent Classification (IPC) or to both national classification and IPC  B. FIELDS SEARCHED  Minimum documentation searched (classification system followed by classification symbols)  U.S.: 210/490, 500.27, 500.35, 500.41, 500.36, 500.42; 264/41, 216; 428/310.5; 96/4, 10, 11; 95/45  Documentation searched other than minimum documentation to the extent that such documents are included in the fields searched				
Electronic data	a base consulted during the international search (name	of data base and, where practicable, sear	ch terms used)	
C. DOCU	MENTS CONSIDERED TO BE RELEVANT			
Category *	Citation of document, with indication, where app	propriate, of the relevant passages	Relevant to claim No.	
Х	US 5.914.039 A (MAHENDRAN et al.) 22 June 1999	, column 3, lines 52-63, column 7,	1-6, 8, 9	
Y	lines 18-33, column 8, lines 21-39, column 8, lines 30 column 11, lines 39-54, claim 1, and column 10, lines	1-34, column 7, second paragraph, 1-45-56)	7	
Y				
Y	lines 5-20 and last paragraph. US 3,676,193 A (COOPER et al) 11 July 1972, claims 1, 8,-11, and 21, entire disclosure.			
			·	
Further	documents are listed in the continuation of Box C.	See patent family annex.		
* S "A" document	pecial categories of cited documents:  t defining the general state of the art which is not considered to be of relevance  plication or patent published on or after the international filing date	"T" later document published after the integrated date and not in conflict with the appliance or theory underlying the investigation."  "X" document of particular relevance; the considered novel or cannot be considered.	eation but cited to understand the ention claimed invention cannot be	
"L" document establish specified)	t which may throw doubts on priority claim(s) or which is cited to the publication date of another citation or other special reason (as	when the document is taken alone  "Y" document of particular relevance; the considered to involve an inventive stey with one or more other such documents.	claimed invention cannot be when the document is combined	
ĺ	t referring to an oral disclosure, use, exhibition or other means	obvious to a person skilled in the art		
priority d	t published prior to the international filing date but later than the late claimed	"&" document member of the same patent		
1	ctual completion of the international search	Date of mailing of the international sear	ren report	
Name and m Ma Con P.C Ale	er 2005 (28.09.2005) ailing address of the ISA/US il Stop PCT, Attn: ISA/US mmissioner for Patents b. Box 1450 c. (703.305.3230	Authorized officer Ana M. Fortuna Telephone No. (571) 272-1900	thifte	

## PATENT COOPERATION TREATY

From the INTERNATI	IONAL SEARCH	ING AUTHO	ORITY				
To: JIANG JI 4 HAWK DRIVE		PCT					
SALEM, NH 03079		WRITTEN OPINION OF THE INTERNATIONAL SEARCHING AUTHORITY					
						(PCT Rule 43bis.1)	
					Date of mailing (day/month/year)	24 OCT 2008	
	s or agent's file re	ference			FOR FURTHER A	ACTION See paragraph 2 below	
000037663	al application No.		Internation	onal filing date	(day/month/year)	Priority date (day/month/year)	$\dashv$
PCT/US04				2004 (14.07.200		16 July 2003 (16.07.2003)	
	al Patent Classific	cation (IPC)				10 341) 2003 (10.07.2003)	
IPC(7): BC	O1D 61/00, 63/00	and US Cl.: 2	210/490, 5	00.27, 500.35,	500.41, 500.36, 500.42	2; 264/41, 216; 428/310.5; 96/4, 10, 11; 95/4	15
JI, JIANG							
1. This o	pinion contains in	dications rela	ating to the	e following iten	18:		
	Box No. I	Basis of the	opinion				
	Box No. II	Priority					
	Box No. III	Non-establi	shment of	opinion with re	gard to novelty, inver	ntive step and industrial applicability	
	Box No. IV	IV Lack of unity of invention					
	Box No. V	Reasoned statement under Rule 43bis.1(a)(i) with regard to novelty, inventive step or industrial applicability; citations and explanations supporting such statement					
	Box No. VI	Certain doc	Certain documents cited				
	Box No. VII	Certain defects in the international application					
	Box No. VIII						
	THER ACTIO					•	
Intern	national Prelimina	ary Examinin his one to be	ng Author the IPEA	ity ("IPEA") e and the chosen	except that this does	be considered to be a written opinion of t not apply where the applicant chooses as International Bureau under Rule 66.1 bis ared.	an
IPFA	a written reply to	ngether, when	e appropri	iate, with amen	dments, before the ex	PEA, the applicant is invited to submit to to piration of 3 months from the date of maili whichever expires later.	the .ng
For further options, see Form PCT/ISA/220.							
3. For further details, see notes to Form PCT/ISA/220.							
Name and	mailing address	of the ISA/ U	IS	Date of comp	letion of this opinion	Authorized officer	
Mait Stop PCT, Attn: ISA/US Commissioner for Patents 28 September			2005 (28.09.2005)	Ana M. Fortuna	Pd		
P.O. Box 1450 Alexandria, Virginia 22313-1450			Telephone No. (571) 272-1700	1			

Facsimile No. (703) 305-3230
Form PCT/ISA/237 (cover sheet) (April 2005)

## WRITTEN OPINION OF THE INTERNATIONAL SEARCHING AUTHORITY

International application No.

PCT/US04/22502

Box No. I Basis of this opinion			
·			
1. With regard to the language, this opinion has been established on the basis of:			
the international application in the language in which it was filed			
a translation of the international application into, which is the language of a translation furnished for the purposes of international search (Rules 12.3(a) and 23.1(b)).			
2. With regard to any nucleotide and/or amino acid sequence disclosed in the international application and necessary to the claimed invention, this opinion has been established on the basis of:			
a. type of material			
a sequence listing			
table(s) related to the sequence listing			
b. format of material			
on paper			
in electronic form			
c. time of filing/furnishing			
contained in the international application as filed.			
filed together with the international application in electronic form.			
furnished subsequently to this Authority for the purposes of search.			
In addition, in the case that more than one version or copy of a sequence listing and/or table(s) relating thereto has been filed or furnished, the required statements that the information in the subsequent or additional copies is identical to that in the application as filed or does not go beyond the application as filed, as appropriate, were furnished.			
4. Additional comments:			

## WRITTEN OPINION OF THE INTERNATIONAL SEARCHING AUTHORITY

International application No.

PCT/US04/22502

Box No. IV Lack of unity of invention				
In response to the invitation (Form PCT/ISA/206) to pay additional fees the applicant has, within the applicable time limit:  paid additional fees  paid additional fees under protest and, where applicable, the protest fee  paid additional fees under protest but the applicable protest fee was not paid  not paid additional fees				
2. This Authority found that the requirement of unity of invention is not complied with and chose not to invite the applicant to				
pay additional fees.  3. This Authority considers that the requirement of unity of invention in accordance with Rule 13.1, 13.2 and 13.3 is				
complied with				
not complied with for the following reasons:				
See the lack of unity section of the International Search Report(Form PCT/ISA/210)				
4. Consequently, this opinion has been established in respect of the following parts of the international application:				
all parts.				
the parts relating to claims Nos. 1-9				
<u>.</u>				

## WRITTEN OPINION OF THE INTERNATIONAL SEARCHING AUTHORITY

International application No. PCT/US04/22502

Box No. V Reasoned statement under Rule 43 bis.1(a)(i) with regard to novelty, inventive step or industrial applicability; citations and explanations supporting such statement

1. Statement		
Novelty (N)	Claims 7	YES
	Claims <u>1-6, 8-9</u>	NO
Inventive step (IS)	Claims NONE	YES
• • •	Claims 1-9	NO
Industrial applicability (IA)	Claims NONE	YES
	Claims NONE	NO

#### 2. Citations and explanations:

Claims 1-6, 8-9 novelty under PCT Article 33(2) as being anticipated by

Mahendran et al (US 5,914,039) (hereinafter '039).

Patent '039 discloses a membrane including the elements (i)-(iii) as claimed in claim 1 (Fig. 1, elements 39, 39', 37, and 38, column 3, lines 52-63, column 7, lines 18-33, column 8, lines 21-39).

Claim 2 is also disclosed (column 8, lines 30-34).

Re: claim 3, the middle layer is formed from organic polymer, e.g. hydrophilic PVDF (column 7, second paragraph).

Limitations of claims 4-6 are further disclosed in patent '039 (see column 7, lines 1-36)

Re: claims 4-6 and 8, the membrane in tubular or hollow fiber shape is disclosed (column 1, lines 11-26, column 11, lines 39-54, claim

Re: claim 9, the claimed membrane properties are disclosed in column 10, lines 45-56).

Claim 7 lacks an inventive step under PCT Article 33(3) as being obvious over Mahendran et al (5,914,039)(hereinafter 'patent 039) as applied to claim 1 above, and further in view of Strobel et al (5, 766, 473)(hereinafter patent '473).

Patent '039 discloses film formation of the claimed membrane composition, coating spheres or particles with the particular support is not disclosed. It would have been obvious to one skilled in the art at the time the invention was made to use any support, e.g. flat, tubular, or particles for the membrane depending on the intended use, spherical membrane or particles will be suitable for example infiltration in packed columns or in chromatographic separations. Patent '473 teaches hydrophilic porous particles or spheres including a substrate and a coating of hydrophilic membrane (abstract, column 4, lines 5-20 and last paragraph); the substrate materials are also disclosed (column 5, lines 5-61). It would have been obvious to one skilled in the art at the time the invention was made to use a polymeric substrate with spherical shape, as disclosed in '473), to support the membrane of '039, depending on the desire configuration or intended use. As to claim 11, other configurations, such as tubules (e.g. flat or oval shape tube, it would have been obvious to one skilled in the art at the time the invention was made, and can be ached by shaping or molding the polymeric support before coating, since the support is flexible, modifications of the membrane diameter shape can be within the knowledge of the skilled artisan.

Claims 2 and 4 lack an inventive step under PCT Article 33(3) as being obvious over Mahendran et al (5,914,039) (hereinafter 'patent 039) as applied to claim 1 above, and further in view of Cooper et al (3,676,193) (hereinafter patent '193). Patent '039 fails to disclose the coating solutions of different polymers, as claimed in a second embodiment of claim 2. Patent '193 teaches a braided support impregnated and coated with a first polymer, coating the support to form a layers on the braided polymeric material, and further using the coated support to canst a film of an additional polymeric membrane, to increase the thickness of the membrane is disclosed (seed entire disclosure, in particular claims 1, 8-11, and 21). Based on the discussed teachings, it would have been obvious to one skilled in the art wishing to have a composite membrane on a braided support with distinct layers of polymer, to use the treated support of '193, which contains a layer of polymer, e.g. polyepoxy, polyurethane, etc (column 8, second paragraph), as support, and further cast the membrane with the hydrophilic membrane of '039, e.g. to provide strength to the membrane, and inherent (as produced by epoxy polymer, as claimed in claim 6) easier binding between the hydrophilic.

# CHAPTER I PCT TELEPHONE MEMORANDUM FOR LACK OF UNITY OF INVENTION



OCT 11 DCT/1100.4	(22522	
PCT No.: PCT/US04/	722502	
Examiner: Ana M. F	<del>f</del> ortuna	
Attorney spoken to:	JI JIANG	•
Date of call: 28 Sep	otember 2005	
Amour	nt of payment approved:	
Deposi	it account number to be charged:	
Attom	ey elected to pay for <u>ALL</u> additional inventions	·
Attorn	ey elected to pay only for the additional inventions cov	vered by
	Group(s):	
encompa	ssing –	
	Claim(s):	·
Attom (Group	ey elected NOT to pay for any additional inventions, to I) covered by Claim(s) <u>1-9</u> has been searched.	herefore, only the first claimed invention
Attorn	ey was orally advised that there is no right to protest	for any group not paid for.
	ley was orally advised that any protest must be filed n Search Report (PCT/ISA/210).	o later than <u>1 month</u> from the mailing

#### **Time Limit For Filing A Protest**

Applicant is hereby given <u>1 month</u> from the mailing date of this Search Report in which to file a protest of the holding of lack of unity of invention. In accordance with PCT Rule 40.2, applicant may protest the holding of lack of unity only with respect to the group(s) paid for.

## **Detailed Reasons For Holding Lack of Unity of Invention:**

Please See Continuation Sheet

Note: A copy of this form must be attached to the Search Report.

International application No: PCT/US04/22502

## ATTACHMENT TO CHAPTER I PCT TELEPHONE MEMORANDUM FOR LACK OF UNITY OF INVENTION

#### Continuation of Detailed Reasons For Holding Lack of Unity of Invention:

This application contains the following inventions or groups of inventions, which are not so linked as to form a single general inventive concept under PCT Rule 13.1. In order for all inventions to be examined, the appropriate additional examination fees must be paid.

Group I claim(s) 1-9, drawn to a composite membrane.

Group II, claim(s) s 10-14, drawn to a method of making a membrane.

Group IL claim(s) 15, drawn to an apparatus.

Group IV, claim(s) 18, drawn to a process of using the membrane.

The inventions listed as Groups I, II, III, and IV do not relate to a single general inventive concept under PCT Rule 13.1 because, under PCT Rule 13.2, they lack the same or corresponding special technical features for the following reasons: the membrane of group I lack the composition, the control of the coating thickness, the speed of collection and the removal of additive recited in claims of group II. Group II includes the feature "spinneret" and "multiple inlets, which features are not resent or required in the claims of groups I, II, or IV. Group IV does not include any common feature with membrane of group I, the process of making the membrane, or the apparatus of group II, and is just directed to using a membrane for multiple fluid separation alternatively.

#### NOTES TO FORM PCT/ISA/220 (continued)

The letter must indicate the differences between the claims as filed and the claims as amended. It must, in particular, indicate, in connection with each claim appearing in the international application (it being understood that identical indications concerning several claims may be grouped), whether

- (i) the claim is unchanged:
- (ii) the claim is cancelled;
- (iii) the claim is new;
- (iv) the claim replaces one or more claims as filed;
- (v) the claim is the result of the division of a claim as filed.

The following examples illustrate the manner in which amendments must be explained in the accompanying letter:

- [Where originally there were 48 claims and after amendment of some claims there are 51]:
   "Claims 1 to 29, 31, 32, 34, 35, 37 to 48 replaced by amended claims bearing the same numbers;
   claims 30, 33 and 36 unchanged; new claims 49 to 51 added."
- 2. [Where ongine lly there were 15 claims and after amendment of all claims there are 11]: "Claims 1 to 15 replaced by amended claims 1 to 11."
- 3. [Where originally there were 14 claims and the amendments consist in cancelling some claims and in adding new claims]:
  "Claims I to 6 and 14 unchanged; claims 7 to 13 cancelled; new claims 15, 16 and 17 added." or
  "Claims 7 to 15 cancelled; new claims 15, 16 and 17 added; all other claims unchanged."
- 4. [Where various kinds of amendments are made]: "Claims 1-10 cmchanged; claims 11 to 13, 18 and 19 cancelled; claims 14, 15 and 16 replaced by amended claim 14; claim 17 subdivided into amended claims 15, 16 and 17; new claims 20 and 21 added."

## "Statement under Article 19(1)" (Rule 46.4)

The amendments may be accompanied by a statement explaining the amendments and indicating any impact that such amendments might have on the description and the drawings (which cannot be amended under Article 19(1)).

The statement will be published with the international application and the amended claims.

## It must be in the Language in which the international application is to be published.

It must be brief, not exceeding 500 words if in English or if translated into English.

It should not be confused with and does not replace the letter indicating the differences between the claims as filed and as amended. It must be filed on a separate sheet and must be identified as such by a heading, preferably by using the words "Statement under Article 19(1)."

It may not contain any disparaging comments on the international search report or the relevance of citations contained in that report. Reference to citations, relevant to a given claim, contained in the international search report may be made only in connection with an amendment of that claim.

## Consequence if a demand for international preliminary examination has already been filed

If, at the time of filing any amendments and any accompanying statement, under Article 19, a demand for international preliminary examination has already been submitted, the applicant must preferably, at the time of filing the amendments (and any statement) with the International Bureau, also file with the International Preliminary Examining Authority a copy of such amendments (and of any statement) and, where required, a translation of such amendments for the procedure before that Authority (see Rules 55.3(a) and 62.2, first sentence). For further information, see the Notes to the demand form (PCT/IPEA/401).

## Consequence with regard to translation of the international application for entry into the national phase

The applicant's amention is drawn to the fact that, upon entry into the national phase, a translation of the claims as amended under Article 19 may have to be furnished to the designated/elected Offices, instead of, or in addition to, the translation of the claims as filed.

For further details on the requirements of each designated elected Office, see the PCT Applicant's Guide, Volume II.